

FORM PTO-1390

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE
TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER:
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U.S. PPN, NO. (if known, see 37 CFR 1.5)
10/018885

INTERNATIONAL APPLICATION NO.:
PCT/FR00/01740

INTERNATIONAL FILING DATE:
22 JUNE 2000

PRIORITY DATE CLAIMED:
23 JUNE 1999

TITLE OF INVENTION: PHYTOSANITARY COMPOSITION COMPRISING AT LEAST A WATER SOLUBLE AGENT AND A MODIFIED OIL

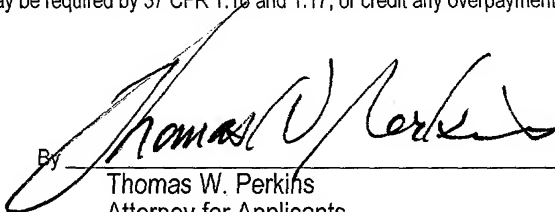
APPLICANT(S) FOR DO/EO/US: Alain MILIUS, Christian GAUVRIT, Thomas MULLER, Bernard BRANCO

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
 2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
 3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
 4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
 5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ has been transmitted by the International Bureau. (see attached copy of PCT/IB/308)
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
 6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
 - ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
 8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
 9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
 10. ☐ A translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).
- Item 11. to 16. below concern document(s) or information included:
11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
 12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
 13. ☒ A **FIRST** preliminary amendment.
 - ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
 14. ☐ A substitute specification.
 15. ☐ A change of power of attorney and/or address letter.
 16. ☒ Other items or information:

International Search Report
PCT/IPEA/409
Application Data Sheet

Abstract of the Disclosure on a Separate Sheet
PCT/IB/308

U.S. APPLICATION NO. (if known, see 37 CFR 1.53) 10/018885		INTERNATIONAL APPLICATION NO. PCT/FR00/01740		ATTORNEY'S DOCKET NO. S 5015 PCT/US	
17. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)): Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$ 1,040.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$ 890.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$ 740.00 International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$ 710.00 International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$ 100.00 <div style="text-align: right;">ENTER APPROPRIATE BASIC FEE AMOUNT =</div>				CALCULATIONS PTO USE ONLY	
Surcharge of \$130.00 for furnishing the oath or declaration later than 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	130.00
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$	
Total claims	14 - 20 =	0	X \$18.00	\$	
Independent claims	2 - 3 =	0	X \$84.00	\$	
MULTIPLE DEPENDENT CLAIMS(S) (if applicable)			+ \$280.00	\$	
TOTAL OF ABOVE CALCULATIONS =				\$	1,020.00
Reduction of 1/2, if applicant is entitled to Small Entity status under 37 CFR 1.27.				+	\$
SUBTOTAL =				\$	1,020.00
Processing fee of \$130 for furnishing the English translation later than 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
TOTAL NATIONAL FEE =				\$	1,020.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				+	\$
TOTAL FEES ENCLOSED =				\$	1,020.00
				Amount to be refunded:	
				charged:	
a. <input checked="" type="checkbox"/> A check in the amount of \$ 1,020.00 to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. 25-0120 in the amount of \$ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required by 37 CFR 1.16 and 1.17, or credit any overpayment to Deposit Account No. 25-0120 . A duplicate copy of this sheet is enclosed.					
SEND ALL CORRESPONDENCE TO: <div style="display: flex; justify-content: space-between;"> <div> CUSTOMER NO. 00466 YOUNG & THOMPSON 745 South 23rd Street 2nd Floor Arlington, VA 22202 (703) 521-2297 facsimile (703) 685-0573 </div> <div> December 26, 2001 </div> <div style="text-align: right;"> By  Thomas W. Perkins Attorney for Applicants Registration No. 33,027 </div> </div>					

PATENTS

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

Alain MILIUS et al.

Serial No. (unknown)

Filed herewith

PHYTOSANITARY COMPOSITION
COMPRISING AT LEAST A WATER
SOLUBLE AGENT AND A MODIFIED OIL

PRELIMINARY AMENDMENT

Commissioner for Patents

Washington, D.C. 20231

Sir:

Prior to calculation of the filing fee, please amend
the above-identified application as follows:

IN THE CLAIMS:

Amend the claims as follows:

--3. (amended) The method such as defined in claim
1, in which the composition employed comprises a mixture of a
number of modified oils, and more particularly a mixture of
one or more ethoxylated oils, with one or more ethoxylated
esters of oils.--

--4. (amended) The method such as defined in claim
1, in which the modified oil comprised in the composition
employed is an ethoxylated methyl ester of an oil having an EO
index of greater than or equal to 8 and less than or equal to
15.--

--5. (amended) The method such as defined in claim 1, in which the modified oil comprised in the composition employed is an ethoxylated oil having an EO index of greater than or equal to 30 and less than or equal to 40.--

--6. (amended) The method such as defined in claim 1, in which the modified oil employed is modified rapeseed oil or modified sunflower oil.--

--7. (amended) The method such as defined in claim 1, in which the phytosanitary active principle is a compound of chemical structure derived from the radical:

$-C(=O)-CH_2-N-CH_2-P(=O)$ and more particularly glyphosate or N-(phosphonomethyl)glycine, in the form of a water-soluble salt, such as, for example, the monoisopropylamine or trimethylsulfonium salts.--

--10. (amended) The composition such as defined in claim 8, in which the active principle is a compound of chemical structure derived from the radical $-C(=O)-CH_2-N-CH_2-P(=O)$, and is very particularly glyphosate or N-phosphonomethylglycine, in the form of a water-soluble salt, such as, for example, the monoisopropylamine or trimethylsulfonium salts.--

--11. (amended) The composition such as defined in claim 8, comprising an alkoxyated vegetable oil and an alkoxyated alkyl ester of a vegetable oil.--

--14. (amended) The composition such as defined in claim 12, in which the modified oil employed is modified rapeseed oil or modified sunflower oil.--

R E M A R K S

The above changes in the claims merely place this national phase application in the same condition as it was during the international phase, with the multiple dependencies being removed.

Attached hereto is a marked-up version of the changes made to the claims by the current amendment. The attached page is captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE".

Respectfully submitted,

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By 

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December 26, 2001

VERSION WITH MARKINGS TO SHOW CHANGES MADE

The claims have been amended as follows:

3. (amended) The method such as defined in ~~one of~~
~~claimsclaim 1-or-2~~, in which the composition employed com-
prises a mixture of a number of modified oils, and more
particularly a mixture of one or more ethoxylated oils, with
one or more ethoxylated esters of oils.

4. (amended) The method such as defined in ~~one of~~
~~claimsclaim 1-or-2~~, in which the modified oil comprised in the
composition employed is an ethoxylated methyl ester of an oil
having an EO index of greater than or equal to 8 and less than
or equal to 15.

5. (amended) The method such as defined in ~~one of~~
~~claimsclaim 1-or-2~~, in which the modified oil comprised in the
composition employed is an ethoxylated oil having an EO index
of greater than or equal to 30 and less than or equal to 40.

6. (amended) The method such as defined in ~~one of~~
~~claimsclaim 1-to-5~~, in which the modified oil employed is
modified rapeseed oil or modified sunflower oil.

7. (amended) The method such as defined in ~~one of~~
~~claimsclaim 1-to-6~~, in which the phytosanitary active princi-
ple is a compound of chemical structure derived from the
radical: $-C(=O)-CH_2-N-CH_2-P(=O)$ and more particularly
glyphosate or N-(phosphonomethyl)glycine, in the form of a
water-soluble salt, such as, for example, the
monoisopropylamine or trimethylsulfonium salts.

10. (amended) The composition such as defined in ~~one of claims~~claim 8 ~~or 9~~, in which the active principle is a compound of chemical structure derived from the radical - $C(=O)-CH_2-N-CH_2-P(=O)$, and is very particularly glyphosate or N-phosphonomethylglycine, in the form of a water-soluble salt, such as, for example, the monoisopropylamine or trimethylsulfonium salts.

11. (amended) The composition such as defined in ~~one of claims~~claim 8 ~~to 10~~, comprising an alkoxyated vegetable oil and an alkoxyated alkyl ester of a vegetable oil.

14. (amended) The composition such as defined in ~~one of claims~~claim 12 ~~or 13~~, in which the modified oil employed is modified rapeseed oil or modified sunflower oil.

20. 21. 22. 23. 24. 25. 26. 27. 28. 29. 30. 31. 32. 33. 34. 35. 36. 37. 38. 39. 40. 41. 42. 43. 44. 45. 46. 47. 48. 49. 50. 51. 52. 53. 54. 55. 56. 57. 58. 59. 60. 61. 62. 63. 64. 65. 66. 67. 68. 69. 70. 71. 72. 73. 74. 75. 76. 77. 78. 79. 80. 81. 82. 83. 84. 85. 86. 87. 88. 89. 90. 91. 92. 93. 94. 95. 96. 97. 98. 99. 100. 101. 102. 103. 104. 105. 106. 107. 108. 109. 110. 111. 112. 113. 114. 115. 116. 117. 118. 119. 120. 121. 122. 123. 124. 125. 126. 127. 128. 129. 130. 131. 132. 133. 134. 135. 136. 137. 138. 139. 140. 141. 142. 143. 144. 145. 146. 147. 148. 149. 150. 151. 152. 153. 154. 155. 156. 157. 158. 159. 160. 161. 162. 163. 164. 165. 166. 167. 168. 169. 170. 171. 172. 173. 174. 175. 176. 177. 178. 179. 180. 181. 182. 183. 184. 185. 186. 187. 188. 189. 190. 191. 192. 193. 194. 195. 196. 197. 198. 199. 200. 201. 202. 203. 204. 205. 206. 207. 208. 209. 210. 211. 212. 213. 214. 215. 216. 217. 218. 219. 220. 221. 222. 223. 224. 225. 226. 227. 228. 229. 230. 231. 232. 233. 234. 235. 236. 237. 238. 239. 240. 241. 242. 243. 244. 245. 246. 247. 248. 249. 250. 251. 252. 253. 254. 255. 256. 257. 258. 259. 260. 261. 262. 263. 264. 265. 266. 267. 268. 269. 270. 271. 272. 273. 274. 275. 276. 277. 278. 279. 280. 281. 282. 283. 284. 285. 286. 287. 288. 289. 290. 291. 292. 293. 294. 295. 296. 297. 298. 299. 300. 301. 302. 303. 304. 305. 306. 307. 308. 309. 310. 311. 312. 313. 314. 315. 316. 317. 318. 319. 320. 321. 322. 323. 324. 325. 326. 327. 328. 329. 330. 331. 332. 333. 334. 335. 336. 337. 338. 339. 340. 341. 342. 343. 344. 345. 346. 347. 348. 349. 350. 351. 352. 353. 354. 355. 356. 357. 358. 359. 360. 361. 362. 363. 364. 365. 366. 367. 368. 369. 370. 371. 372. 373. 374. 375. 376. 377. 378. 379. 380. 381. 382. 383. 384. 385. 386. 387. 388. 389. 390. 391. 392. 393. 394. 395. 396. 397. 398. 399. 400. 401. 402. 403. 404. 405. 406. 407. 408. 409. 410. 411. 412. 413. 414. 415. 416. 417. 418. 419. 420. 421. 422. 423. 424. 425. 426. 427. 428. 429. 430. 431. 432. 433. 434. 435. 436. 437. 438. 439. 440. 441. 442. 443. 444. 445. 446. 447. 448. 449. 450. 451. 452. 453. 454. 455. 456. 457. 458. 459. 460. 461. 462. 463. 464. 465. 466. 467. 468. 469. 470. 471. 472. 473. 474. 475. 476. 477. 478. 479. 480. 481. 482. 483. 484. 485. 486. 487. 488. 489. 490. 491. 492. 493. 494. 495. 496. 497. 498. 499. 500. 501. 502. 503. 504. 505. 506. 507. 508. 509. 510. 511. 512. 513. 514. 515. 516. 517. 518. 519. 520. 521. 522. 523. 524. 525. 526. 527. 528. 529. 530. 531. 532. 533. 534. 535. 536. 537. 538. 539. 540. 541. 542. 543. 544. 545. 546. 547. 548. 549. 550. 551. 552. 553. 554. 555. 556. 557. 558. 559. 560. 561. 562. 563. 564. 565. 566. 567. 568. 569. 570. 571. 572. 573. 574. 575. 576. 577. 578. 579. 580. 581. 582. 583. 584. 585. 586. 587. 588. 589. 590. 591. 592. 593. 594. 595. 596. 597. 598. 599. 600. 601. 602. 603. 604. 605. 606. 607. 608. 609. 610. 611. 612. 613. 614. 615. 616. 617. 618. 619. 620. 621. 622. 623. 624. 625. 626. 627. 628. 629. 630. 631. 632. 633. 634. 635. 636. 637. 638. 639. 640. 641. 642. 643. 644. 645. 646. 647. 648. 649. 650. 651. 652. 653. 654. 655. 656. 657. 658. 659. 660. 661. 662. 663. 664. 665. 666. 667. 668. 669. 670. 671. 672. 673. 674. 675. 676. 677. 678. 679. 680. 681. 682. 683. 684. 685. 686. 687. 688. 689. 690. 691. 692. 693. 694. 695. 696. 697. 698. 699. 700. 701. 702. 703. 704. 705. 706. 707. 708. 709. 710. 711. 712. 713. 714. 715. 716. 717. 718. 719. 720. 721. 722. 723. 724. 725. 726. 727. 728. 729. 730. 731. 732. 733. 734. 735. 736. 737. 738. 739. 740. 741. 742. 743. 744. 745. 746. 747. 748. 749. 750. 751. 752. 753. 754. 755. 756. 757. 758. 759. 760. 761. 762. 763. 764. 765. 766. 767. 768. 769. 770. 771. 772. 773. 774. 775. 776. 777. 778. 779. 780. 781. 782. 783. 784. 785. 786. 787. 788. 789. 790. 791. 792. 793. 794. 795. 796. 797. 798. 799. 800. 801. 802. 803. 804. 805. 806. 807. 808. 809. 810. 811. 812. 813. 814. 815. 816. 817. 818. 819. 820. 821. 822. 823. 824. 825. 826. 827. 828. 829. 830. 831. 832. 833. 834. 835. 836. 837. 838. 839. 840. 841. 842. 843. 844. 845. 846. 847. 848. 849. 850. 851. 852. 853. 8

A phytosanitary treatment method by foliar absorption that uses a composition with at least a water soluble phytosanitary active principle and at least a modified oil, the composition having at least a phytosanitary active principle, at least a vegetable oil and/or an ethoxylated vegetable oil ester.

- 1 -

PHYTOSANITARY COMPOSITION COMPRISING AT LEAST A WATER SOLUBLE AGENT AND A MODIFIED OIL

5 The invention relates to a method for phytosanitary treatment by foliar absorption, employing a combination between an active principle and a modified oil and a novel combination for employing said method.

10 The penetration of a phytosanitary active principle into a plant takes place either at leaf level, by foliar absorption, or at root level, by radicular absorption. The foliar absorption of an active principle is often difficult and very slow; this is especially the case for glyphosate which must be
15 formulated with surface-active agents to increase this penetration. In the international patent application published under the number WO 99/03343, the inventors propose the use of methyl esters of ethoxylated fatty acids, in combination with vegetable oils, as well as
20 wetting, dispersing and emulsifying agents, in herbicidal concentrations, to improve their stability. However, this approach is not suitable for water-soluble active materials because it leads to emulsions.

25 However, when small quantities of glyphosate are treated, that is to say of less than or equal to 306 grams per hectare, as is the case in the supervision of fallow land or for the treatment of young annual plants, none of the adjuvants currently employed with
30 this active principle allow a rate of absorption of greater than 50% after 72 hours to be achieved. A period of rain following the treatment of a crop thus risks inducing pollution of the site, while a period of sunshine can cause the degradation of the active
35 principle.

The applicants have thus endeavored to perfect a method of phytosanitary treatment by foliar absorption which allows a high absorption of a hydrophilic active

principle by the plant in a short time, while limiting the harm to ecosystems, and using adjuvants which are much less irritant, especially for the eyes, than those used until today.

5

The invention relates to a method of phytosanitary treatment by foliar absorption, characterised in that it employs an aqueous composition comprising at least one water-soluble phytosanitary active principle and at least one modified oil, chosen from the ethoxylated oils having an ethylene oxide number, called EO index below, of greater than or equal to 20 and less than or equal to 60 and more particularly greater than or equal to 30 and less than or equal to 50 or from ethoxylated methyl, ethyl, linear or branched propyl or linear or branched butyl esters of oils, having an EO index of greater than or equal to 5 and less than or equal to 50 and more particularly greater than or equal to 6 and less than or equal to 20.

20

The modified oils used in the context of the present invention are of plant origin or of animal origin. Among the modified oils of animal origin which are employed in the method such as is defined above, is, for example, modified tallow oil. Among the modified oils of plant origin which are employed in the method such as is defined above, are, for example, modified sunflower, linseed, soybean, corn, peanut, copra, olive, palm, hydrogenated palm or rapeseed oils.

30

According to a particular variant of the method, such as defined above, the modified oil comprised in the composition employed is an ethoxylated methyl ester of an oil having an EO index of greater than or equal to 8 and less than or equal to 15.

35

According to another particular variant of the method, such as defined above, the modified oil comprised in the composition employed is an ethoxylated oil having

an EO index of greater than or equal to 30 and less than or equal to 40.

5 The invention particularly relates to a method such as defined above, for which the modified oil comprised in the composition employed is modified rapeseed oil or modified sunflower oil.

10 By at least one modified oil, it is indicated that the composition employed in the method which is the subject of the present invention can comprise either a single modified oil, or a mixture of modified oils; in this latter case, it can be a mixture of modified oils of the same origin or a mixture of modified oils of
15 different origins; it can also be a mixture of one or more ethoxylated oils such as are defined above with one or more ethoxylated esters of oils such as are defined above.

20 Phytosanitary treatment is understood in the context of the present invention as preferably meaning a fungicidal, insecticidal or herbicidal treatment and more particularly a treatment in which the active principle is a compound of chemical structure derived
25 from the radical: $-C(=O)-CH_2-N-CH_2-P(=O)$.

30 An example of a compound of chemical structure derived from said radical is more particularly glyphosate or N-(phosphonomethyl)glycine, in the form of a water-soluble salt, such as, for example, the monoisopropylamine or trimethylsulfonium salts.

35 According to another aspect of the present invention, this relates to a composition comprising at least one water-soluble phytosanitary active principle, and at least one modified vegetable oil, characterized in that said modified vegetable oil is chosen from the ethoxylated oils having an ethylene oxide number of greater than or equal to 20 and less than or equal to

60 and more particularly greater than or equal to 30 and less than or equal to 50 or from the ethoxylated methyl, ethyl, linear or branched propyl or linear or branched butyl esters of oils, having an EO index of greater than or equal to 5 and less than or equal to 50 and more particularly greater than or equal to 6 and less than or equal to 20.

In order to improve its behavior in the cold, the alkoxyated oil can be prepared by incorporating, before its alkoxylation, from 1% to 10% by weight of glycerol. This problem is likewise resolved, by combining in the same composition, an alkoxyated vegetable oil such as defined above and an alkoxyated alkyl ester of a vegetable oil such as defined above, and more particularly an ethoxylated methyl, ethyl, linear or branched propyl or linear or branched butyl ester of a vegetable oil.

The invention relates more particularly to a composition, such as defined above, in which the active principle is a compound of chemical structure derived from the radical $-C(=O)-CH_2-N-CH_2-P(=O)$, and is very particularly glyphosate or N-phosphonomethylglycine, in the form of a water-soluble salt, such as, for example, the monoisopropylamine or trimethylsulfonium salts.

The invention relates very particularly to a composition such as defined above, in which the modified oil is an ethoxylated methyl ester of an oil having an EO index of greater than or equal to 8 and less than or equal to 15 and the phytosanitary active principle is glyphosate in the form of a water-soluble salt.

35

The invention also relates very particularly to a composition such as defined above, in which the modified oil is an ethoxylated oil having an EO index of greater than or equal to 30 and less than or equal

to 40 and the phytosanitary active principle is glyphosate in the form of a water-soluble salt.

The invention finally relates very particularly to a composition such as defined above, in which the modified oil is modified rapeseed oil or modified sunflower oil.

EXAMPLE 1: PREPARATION OF MODIFIED OILS

A) Preparation of ethoxylated methyl esters of rapeseed oil

The ethoxylated methyl esters of rapeseed oil are obtained from the methyl ester of rapeseed oil by reaction for approximately 45 minutes, at 180°C and under a pressure of 4.5 bar, with the quantity of ethylene oxide necessary for the obtainment of the desired molar ratio, in the presence of a basic catalyst, then cooling and neutralization of the catalyst.

B) Preparation of ethoxylated rapeseed oils

By employing the method of ethoxylation described in the preceding paragraph, in the presence of 2% by weight of glycerol based on the rapeseed oil, the ethoxylated rapeseed oils are obtained.

EXAMPLE 2: STUDY OF THE CAPACITY OF MODIFIED VEGETABLE OILS TO STIMULATE THE FOLIAR PENETRATION OF A PHYTOSANITARY ACTIVE PRINCIPLE

A) Glyphosate compositions

The penetration into the leaves of barley (Hordeum vulgare 1) of glyphosate, which is a hydrophilic herbicide ($\log P < -3.4$), named

N-(phosphonomethyl)glycine, was compared according to the following working method:

Glyphosate labeled with carbon 14 is dissolved in water (4.5 to 21 millimolar; 30 to 50 Bq ηL^{-1}), in the absence of modified vegetable oil. As a reference, the same quantity of radioactive glyphosate is included in a commercial preparation of glyphosate. Ten drops of 0.4 ηL are applied to the adaxial face of the first barley leaf. At 0, 6, 24 and 72 hours, the product which has not penetrated is washed with 0.5 ml of an acetone/water mixture (1:1, V/V) and the radioactivity is measured by liquid scintillation. The radioactivity present in the treated leaf and in the rest of the plant is determined in the carbon dioxide obtained after combustion of the tissue. The results of three series of analyses which were independent of one another, expressed as the percentage of radioactivity which has penetrated into the plant, with respect to the deposited radioactivity, are shown in the following tables:

Table 1

| | FOLIAR PENETRATION A: | | |
|---|-----------------------|------------|------------|
| COMPOSITION | 06 HOURS | 24 HOURS | 72 HOURS |
| Glyphosate +
Ethomeen T/25 (1%) | 28% | 39% | 60% |
| Glyphosate +
rapeseed oil (6 EO) (1%) | 5% | 25% | 42% |
| Glyphosate +
rapeseed oil (10 EO) (1%) | 8% | 30% | 48% |
| Glyphosate +
rapeseed oil (20 EO) (1%) | 18% | 34% | <u>62%</u> |
| Glyphosate +
rapeseed oil (30 EO) (1%) | 28% | <u>62%</u> | <u>63%</u> |
| Glyphosate +
rapeseed oil (40 EO) (1%) | 34% | <u>70%</u> | <u>65%</u> |

The results of this first table demonstrate the superiority of the ethoxylated rapeseed oil, having an EO index of greater than or equal to 20 and especially from 30, as regards the active principle foliar penetration, with respect to that of known additives.

Table 2

| COMPOSITION | FOLIAR PENETRATION A: | | |
|---|-----------------------|------------|------------|
| | 06 HOURS | 24 HOURS | 72 HOURS |
| Glyphosate +
Ethomeen T/25 (1%) | 32% | 47% | 42% |
| Glyphosate +
Methyl ester of rapeseed
oil (2 EO) (1%) | 18% | 32% | 30% |
| Glyphosate +
Methyl ester of rapeseed
oil (3 EO) (1%) | 14% | 28% | 34% |
| Glyphosate +
Methyl ester of rapeseed
oil (4 EO) (1%) | 14% | 34% | 25% |
| Glyphosate +
Methyl ester of rapeseed
oil (6 EO) (1%) | 25% | <u>63%</u> | <u>63%</u> |
| Glyphosate +
Methyl ester of rapeseed
oil (8 EO) (1%) | 41% | <u>60%</u> | <u>73%</u> |

The results of this second table demonstrate the superiority of ethoxylated methyl esters of rapeseed oil having an EO index of greater than or equal to 6 and more particularly of greater than or equal to 8, as regards the foliar penetration of the active principle, with respect to that of known additives.

Table 3

| COMPOSITION | FOLIAR PENETRATION A: | | |
|--------------|-----------------------|----------|----------|
| | 06 HOURS | 24 HOURS | 72 HOURS |
| Glyphosate + | 40% | 50% | 70% |

| | | | |
|---|------------|------------|------------|
| Ethomeen T/25 (1%) | | | |
| Glyphosate + Ethomeen
T/25 (1%) +
Methyl ester of rapeseed
oil (8 EO) (1%) | 60% | <u>80%</u> | <u>85%</u> |
| Glyphosate + Ethomeen
T/25 (1%) +
rapeseed oil (40 EO) (1%) | 49% | 50% | 70% |
| Glyphosate + Ethomeen
T/25 (1%) +
Methyl ester of rapeseed
oil (8 EO) (1%) | <u>46%</u> | <u>50%</u> | <u>74%</u> |
| Glyphosate +
rapeseed oil (40 EO) (1%) | 40% | 50% | 70% |

5 The results of this third table demonstrate the interest that there is in combining one or more modified oils with a hydrophilic active principle for a phytosanitary treatment by foliar absorption.

B) Comparison with the commercial formulations of glyphosate

10 It has been confirmed that the foliar penetration of the isopropylamine salt of the glyphosate with the methyl ester of rapeseed oil (> 8 EO) was more rapid than that of the commercial products based on glyphosate known today, namely Round up™, Round up™
15 Bioforce, Ouragan™ or Sting™ST.

CLAIMS

1. A method of phytosanitary treatment by foliar absorption, characterized in that it employs an aqueous composition comprising at least one water-soluble phytosanitary active principle and at least one modified oil, chosen from the ethoxylated oils having an EO index of greater than or equal to 20 and less than or equal to 60 and more particularly greater than or equal to 30 and less than or equal to 50 or from the ethoxylated methyl, ethyl, linear or branched propyl or linear or branched butyl esters of oils, having an EO index of greater than or equal to 5 and less than or equal to 50 and more particularly greater than or equal to 6 and less than or equal to 20.
2. The method such as defined in claim 1, in which the composition employed comprises at least one or more modified oils of plant origin, chosen from the modified sunflower, linseed, soybean, corn, peanut, copra, olive, palm, hydrogenated palm or rapeseed oils.
3. The method such as defined in one of claims 1 or 2, in which the composition employed comprises a mixture of a number of modified oils, and more particularly a mixture of one or more ethoxylated oils, with one or more ethoxylated esters of oils.
4. The method such as defined in one of claims 1 or 2, in which the modified oil comprised in the composition employed is an ethoxylated methyl ester of an oil having an EO index of greater than or equal to 8 and less than or equal to 15.
5. The method such as defined in one of claims 1 or 2, in which the modified oil comprised in the

composition employed is an ethoxylated oil having an EO index of greater than or equal to 30 and less than or equal to 40.

- 5 6. The method such as defined in one of claims 1 to 5, in which the modified oil employed is modified rapeseed oil or modified sunflower oil.
- 10 7. The method such as defined in one of claims 1 to 6, in which the phytosanitary active principle is a compound of chemical structure derived from the radical: $-C(=O)-CH_2-N-CH_2-P(=O)$ and more particularly glyphosate or N-(phosphonomethyl)glycine, in the form of a water-
15 soluble salt, such as, for example, the monoisopropylamine or trimethylsulfonium salts.
- 20 8. A composition comprising at least one water-soluble phytosanitary active principle, and at least one modified vegetable oil, characterized in that said modified vegetable oil is chosen from the ethoxylated oils having an ethylene oxide number of greater than or equal to 20 and less than or equal to 60 and more particularly greater
25 than or equal to 30 and less than or equal to 50, or from the ethoxylated methyl, ethyl, linear or branched propyl or linear or branched butyl esters of oils having an EO index of greater than or equal to 5 and less than or equal to 50 and more
30 particularly greater than or equal to 6 and less than or equal to 20.
- 35 9. The composition such as defined in claim 8, in which the modified oil of plant origin is chosen from modified sunflower, linseed, soybean, corn, peanut, corpra, olive, palm, hydrogenated palm or rapeseed oils.

10. The composition such as defined in one of claims 8 or 9, in which the active principle is a compound of chemical structure derived from the radical $-C(=O)-CH_2-N-CH_2-P(=O)$, and is very particularly glyphosate or N-phosphonomethylglycine, in the form of a water-soluble salt, such as, for example, the monoisopropylamine or trimethylsulfonium salts.
11. The composition such as defined in one of claims 8 to 10, comprising an alkoxyated vegetable oil and an alkoxyated alkyl ester of a vegetable oil.
12. The composition such as defined in claim 10, in which the modified oil employed is an ethoxyated methyl ester of oil having an EO index of greater than or equal to 8 and less than or equal to 15 and the phytosanitary active principle is glyphosate in the form of a water-soluble salt.
13. The composition such as defined in claim 10, in which the modified oil employed is an ethoxyated oil having an EO index of greater than or equal to 30 and less than or equal to 40 and the phytosanitary active principle is glyphosate in the form of a water-soluble salt.
14. The composition such as defined in one of claims 12 or 13, in which the modified oil employed is modified rapeseed oil or modified sunflower oil.

COMBINED DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that :

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

PHYTOSANITARY COMPOSITION COMPRISING AT LEAST A WATER SOLUBLE AGENT AND A MODIFIED OIL

the specification of which: *(check one)*

REGULAR OR DESIGN APPLICATION

☐ is attached hereto.

☐ was filed on _____ as application Serial No. _____ and was amended on _____ (if applicable).

PCT FILED APPLICATION ENTERING NATIONAL STAGE

☒ was described and claimed in International application No. _____ PCT/FR00/01740 _____ filed on June 22, 2000 and as amended on (if any).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Sec. 1.56.

PRIORITY CLAIM

I hereby claim foreign priority benefits under 35 USC 119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed.

PRIOR FOREIGN APPLICATION(S)

| Country | Application Number | Date of Filing
(day, month, year) | Priority Claimed |
|---------|--------------------|--------------------------------------|------------------|
| FRANCE | 99/08,010 | 23 June 1999 | Yes |
| | | | |

I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional patent application(s) listed below:

(Application Serial No.)

(Filing Date)

Status (patented, pending, abandoned)

(Complete this part only if this is a continuing application.)

I hereby claim the benefit under 35 USC 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of 35 USC 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations Sec. 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)

(Filing Date)

(Status--patented, pending, abandoned)

POWER OF ATTORNEY

The undersigned hereby authorizes the U.S. attorney or agent named herein to accept and follow instructions from **L'AIR LIQUIDE** as to any action to be taken in the Patent and Trademark Office regarding this application without direct communication between the U.S. attorney or agent and the undersigned. In the event of a change in the persons from whom instructions may be taken, the U.S. attorney or agent named herein will be so notified by the undersigned.

As a named inventor, I hereby appoint the registered patent attorneys represented by Customer No. **000466** to prosecute this application and transact all business in the Patent and Trademark Office connected therewith, including: **Robert J. PATCH, Reg. No. 17,355, Andrew J. PATCH, Reg. No. 32,925, Robert F. HARGEST, Reg. No. 25,590, Benoît CASTEL, Reg. No. 35,041, Eric JENSEN, Reg. No. 37,855, Thomas W. PERKINS, Reg. No. 33,027, and Roland E. LONG, Jr., Reg. No. 41,949, c/o YOUNG & THOMPSON, Second Floor, 745 South 23rd Street, Arlington, Virginia 22202.**

Address all telephone calls to Young & Thompson at 703/521-2297. Telefax: 703/685-0573.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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